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Impulsive Stimulated X-ray Raman Scattering in Molecular Systems[1] JORDAN O'NEAL, Stanford University; Stanford PULSE Institute, SLAC National Accelerator Laboratory, SOLÈNE OBERLI, ANTONIO PICÓN, Departamento de Química, Universidad Autónoma de Madrid, 28049 Madrid, Spain, ELIO CHAMPENOIS, JAMES CRYAN, Stanford PULSE Institute, SLAC National Accelerator Laboratory, LS05 COLLABORATION — The field of nonlinear X-ray interactions is growing with technological advances at free-electron lasers. The development of intense attosecond X-ray pulses has opened a new regime for studying nonlinear X-ray interactions in the impulsive limit. In an experiment conducted at the Linac Coherent Light Source, we made the first observation of impulsive stimulated X-ray Raman scattering (ISXRS) in a molecular system, finding a two-photon cross section of $(3 \pm 2) \times 10^{-55}$ cm^{4}s/photon. ISXRS is a versatile process for studying electronic charge motion and X-ray-matter interactions. Simulations matching the observed signal show that ISXRS coherently populates multiple valence-excited electronic states. This is a significant step toward direct observation of coherent electronic wavepackets using the planned 2-color attosecond X-ray mode of the LCLS.

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